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AFRL-SR-BL-TR-01-

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1. REPORT DATE (DD-MM-YYYY) 05-09-2001		2. REPORT TYPE Final Technical		3. DATES COVERED (From - To) 01-03-1998 - 31-05-2001	
4. TITLE AND SUBTITLE (U) Study of the Sub- and Supercritical Behavior of Fuel Droplets				5a. CONTRACT NUMBER	
				5b. GRANT NUMBER F49620-98-1-0373	
				5c. PROGRAM ELEMENT NUMBER 61102	
6. AUTHOR(S) Yaw D. Yeboah				5d. PROJECT NUMBER 2309	
				5e. TASK NUMBER BX	
				5f. WORK UNIT NUMBER	
7. PERFORMING ORGANIZATION NAME(S) AND ADDRESS(ES) Clark Atlanta University 223 James P. Brawley Drive, S.W. Atlanta GA 30314				8. PERFORMING ORGANIZATION REPORT NUMBER	
9. SPONSORING / MONITORING AGENCY NAME(S) AND ADDRESS(ES) AFOSR/NA 801 North Randolph Street Room 732 Arlington VA 22203-1977				10. SPONSOR/MONITOR'S ACRONYM(S)	
				11. SPONSOR/MONITOR'S REPORT NUMBER(S)	
12. DISTRIBUTION / AVAILABILITY STATEMENT Approved for public release; distribution is unlimited					
13. SUPPLEMENTARY NOTES					
14. ABSTRACT A study of the droplet gasification and combustion characteristics of hydrocarbon fuel droplets was conducted at sub- and supercritical. The experimental setup provided quiescent and convective environments under supercritical pressure and temperature conditions. The droplet combustion experiment hardware consisted of: a liquid pressurizing and transfer system; a high pressure and temperature combustion chamber; a droplet formation, deployment and ignition system; and a high speed CCD imaging system. The gasification and combustion characteristics of droplets of several hydrocarbons under quiescent environments were studied. Specifically, the mass emission rates and combustion characteristics of 1.5-mm-diameter suspended droplets of a series of hydrocarbons ranging from hexane (C6) to tetradecane (C14) were measured at sub- and supercritical pressures and temperatures using the high-speed image system. The images of droplet gasification and combustion revealed interesting phenomena and provided better understanding of the droplet behavior in sub- and supercritical pressure and temperature environments.					
15. SUBJECT TERMS supercritical droplet hydrocarbon					
16. SECURITY CLASSIFICATION OF:			17. LIMITATION OF ABSTRACT  UL	18. NUMBER OF PAGES  57	19a. NAME OF RESPONSIBLE PERSON Julian M. Tishkoff
a. REPORT Unclassified	b. ABSTRACT Unclassified	c. THIS PAGE Unclassified			19b. TELEPHONE NUMBER (include area code) (703) 696-8478

20011012 032

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**STUDY OF THE SUB- AND SUPERCRITICAL BEHAVIOR  
OF FUEL DROPLETS**

SEP - 5 2001

**Final Report**

For the Period March 1, 1998 – June, 2001

For

Contract No. F49620-98-1-0373

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## TABLE OF CONTENTS

	Page
Summary.....	3
Description of the Problem.....	4
Background.....	6
Project Tasks and Accomplishments.....	17
Experimental.....	18
Results and Discussion.....	29
Conclusions.....	46
Personnel Supported.....	48
Publications.....	48
Presentations.....	49
Interactions Related to the Research.....	49
Inventions.....	49
Acknowledgments.....	50
References.....	50

## SUMMARY

A study of the droplet gasification and combustion characteristics of hydrocarbon fuel droplets was conducted at sub- and supercritical conditions using a versatile droplet combustion experimental setup developed at Clark Atlanta University. This setup provides quiescent and convective environments under supercritical pressure and temperature conditions. The droplet combustion experiment hardware consists of a liquid pressurizing and transfer system, a high pressure and temperature combustion chamber, a droplet formation, deployment and ignition system, and a high speed Kodak EktaPro CCD imaging system. The gasification and combustion characteristics of droplets of several hydrocarbons under quiescent environments were studied using the setup. Specifically, the mass emission rates and combustion characteristics of ~1.5-mm-diameter suspended droplets of a series of hydrocarbons ranging from hexane ( $C_6$ ) to tetradecane ( $C_{14}$ ) were measured at sub- and supercritical pressures and temperatures using the high-speed image system. The images of droplet gasification and combustion revealed interesting phenomena and provided better understanding of the droplet behavior in sub- and supercritical pressure and temperature environments.

Results showed that both n-hexane and n-heptane droplets closely exhibit classical  $d^2$ -law gasification behavior at subcritical temperatures and at sub- and supercritical pressures. Results of the high pressure droplet gasification under normal gravity conditions showed that the gasification rate increases non-linearly with increase in pressure (at subcritical pressures) up to a maximum at the critical pressure and then decreases at supercritical pressures. The gasification rate was found to increase non-linearly with increasing temperature below the critical temperatures. The acceleration of free-falling hexane and heptane droplets and the resistance force on the droplets were found to vary linearly with temperature. The oscillatory deformation of a non-combusting droplet was enhanced as the system temperature was increased.

The effect of molecular weight on the gasification and combustion behavior of droplets of hydrocarbon fuels from hexane ( $C_6$ ) to tetradecane ( $C_{14}$ ) was also studied at subcritical conditions. The results showed that the system pressure and molecular weight have a pronounced effect on the gasification and combustion behavior of the fuel

droplets. The oscillatory deformation of a free droplet, the buoyancy effects and flame propagation of a combusting free-droplet were found to be directly influenced by the system pressure and temperature. At the critical pressure of decane, the falling rate of its free-drop droplet reached its maximum. On the other hand, the gasification rate of a droplet proportionally decreased with the molecular weight of the hydrocarbon under system pressures below and above the hydrocarbon critical pressure. As the molecular weight of hydrocarbons and the system pressure increased, the ignition of the free-drop droplets became more difficult, the combustion rates of the droplets decreased, and the flames of the combusting free-drop droplets became less stable. Increasing the system's oxygen partial pressure led to enhanced ignition, combustion rate and flame stability of the free droplets.

The video images of the hexane droplet behavior at temperatures close to the critical temperature showed that the droplet becomes dynamically unstable at temperatures close to 500 °C and disintegrates as a result of the vanishing surface tension as the critical temperature is reached.

This study provided new insights and data to advance the scientific understanding of the mass emission and combustion characteristics of fuel droplets. The project was of significant interest to the Air Force and the aerospace propulsion industry, especially in the areas of liquid propellant rocket propulsion and combustion, airbreathing combustion and liquid fuel droplet and spray combustion dynamics in advanced gas turbines and aircraft engines. The project educated and trained Clark Atlanta University students in the areas of combustion and thermal sciences.

## DESCRIPTION OF THE PROBLEM

The phenomena associated with the evaporation of fuel droplets and the methods of predicting their evaporation rate are important in the analysis of engineering operations involving the process of spray cooling, drying, absorption, deposition, humidification, combustion and rocket propulsion. The physical problem considered in this project is the

transient vaporization of fuel droplets placed in an enclosed chamber with nitrogen test gas under sub and supercritical conditions.

The thermodynamic states in the chamber can be varied from subcritical, critical, to supercritical and vice versa. As a result of the heat transfer from the surrounding gases, the droplets start to heat up and evaporation occurs due to the increased vapor concentration gradient near the surface of the droplets. As the temperature and pressure in the chamber change, the thermodynamic state of the gases surrounding the droplets causes the droplets to experience its subcritical regime and eventually the supercritical regime when the temperature and pressure in the chamber exceed their critical points.

The topic has long been an intensive research area due to its practical implications in liquid propellant rocket motors and diesel engines. The process is extremely complex. When the chamber condition is in the thermodynamic subcritical regime of the liquid droplet, the droplet surface provides a well-defined interfacial boundary. In the analysis of the droplet behavior at this subcritical condition, physical processes in the droplet interior and ambient gases can be treated separately and then matched at the interface by requiring liquid-vapor phase equilibrium and continuities of mass and energy fluxes.

The process becomes significantly different in the supercritical regime where chamber pressure and temperature are well above the thermodynamic critical states of the droplet liquid. The droplet may be continuously heated up with its surface reaching the critical point prior to the end of the droplet lifetime. When this occurs, the sharp distinction between the gas and the liquid disappears. The enthalpy of the vaporization reduces to zero, and no abrupt phase change is involved in the vaporization process. The density and temperature of the entire field of liquid droplet and the ambient gases, as well

as their gradients, vary continuously across the droplet surface. The physical states of these two regimes can be depicted in Figure. 1.

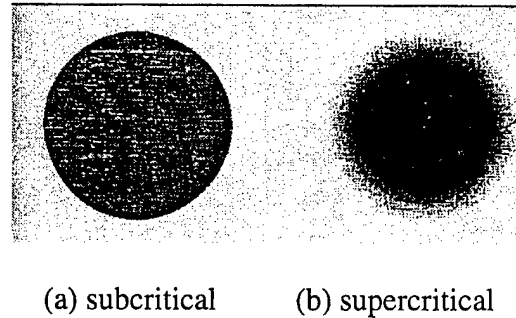


Figure1: Sketch of droplet characteristics in different regimes.

## BACKGROUND

Knowledge of the droplet gasification and combustion processes in high pressure environments and parameters affecting them are needed to achieve proper design and fabrication of internal combustion engines such as diesel, jet or rocket engines. Extensive literature review on both experimental and theoretical research is available on the subject of droplet evaporation and combustion in a stagnant environment or under forced convection conditions at atmospheric or near atmospheric pressures (Bellan, 2000; Shawn and Abraham, 1996). In many industrial applications, droplet evaporation at elevated pressures and temperatures is important, especially when the elevated levels of the pressure and temperature get close to and beyond the critical point. Its potential importance is so well recognized that numerous theoretical and experimental studies aimed at improving the basic understanding of the physics have been pursued. In fact in recent years, diesel engine manufacturers have striven to increase the ambient chamber

density, and thereby the pressure, during fuel injection. This effort is motivated by the understanding that increased ambient density relative to the density of the injected fuel results in better mixing and increased rates of combustion (Abraham et al, 1994; Hou et al, 1994, 1995; and Abraham and Bracco, 1995). As these trends lead to even greater chamber pressures and temperatures, it becomes even more imperative that there is a clear understanding of supercritical processes which would lead to improved vaporizing and combusting spray models.

Research progress in the past decades in the topic area has been reviewed extensively by Faeth (1977), Law (1982), Sirignano (1983), and Chiu (1989). Early experimental investigations include Torda et al. (1968), Faeth et al. (1969), Savery et al. (1970), Matlosz et al. (1972), Canada et al. (1975), and Kadota et al. (1981). Significant work was conducted in the early 20<sup>th</sup> century by Sato et al. (1990), Norton et al. (1990), Jeng et al. (1990), Litchford et al. (1992), Parigger (1994), and Woodward et al. (1994, 1996).

As early as in the 1960's, Faeth et al. (1969) considered the combustion of n-decane droplets in air in the pressure range of 100-20,000 psia and under microgravity conditions. Results indicated a significant change in the burning mechanism at the critical pressure. The droplet-burning lifetime decreased with pressure in the subcritical range, with the entire process dominated primarily by gasification of the droplet. However, further increase of the ambient pressure beyond the critical point caused the combustion mechanism to be diffusion-controlled and increased the burning lifetime. This phenomenon was also found in experiments conducted by Sato et al. (1990).



Matloz et al (1972) measured n-hexane droplets with initial diameter of 04. – 1.65 mm and temperatures of 340 – 360 K vaporizing under normal gravity conditions in quiescent nitrogen gas at reduced temperatures  $T_r = 1.08$  and reduced pressures  $P_r = 0.23, 0.68, 2.72, \text{ and } 3.41$ . They concluded that the portion of the droplet lifetime spent in the  $D^2$ -like vaporization regime decreases as the ambient pressure increases, as does the transient period. They also concluded that the evaporation rate constant for n-hexane increased with increase in ambient pressure for both sub- and supercritical pressure conditions at  $T_r = 1.08$ . Hiroyasu et al (1974) measured the size history and temperature of an n-heptane droplet with initial droplet size of 1.8 mm vaporizing under normal gravity. Their study predicted a non-quasi-steady behavior in supercritical conditions. An increased evaporation rate constant with pressure was reported by Hartfield (1991). Chauveau et al (1993) studied vaporizing droplet size histories under microgravity ( $\sim 10^{-2} g$ ) conditions using high speed film photography and image data processing techniques. Their study on n-heptane droplets indicate that a quasi-steady behavior is followed at ambient pressures and that the evaporation rate constant under microgravity conditions showed a more complicated dependence on pressure. In a review by Shawn et al (1996) it was inferred that for n-pentane through n-dodecane, a vaporizing droplet can reach the critical state for ambient pressures greater than approximately twice the fuel's critical pressure and for the ambient temperatures approximately twice the fuel's critical temperature. Combusting droplets can reach this state when ambient pressure is about 2.5 times that of the fuel's critical pressure.

Recently, Stengele et al (1999) did experimental work on supercritical droplet vaporization and combustion of single component n-pentane and n-nonane droplets and

binary fuel droplets of n-pentane and n-nonane in nitrogen under normal gravity conditions. Detailed measurements were conducted with one- and two-component droplets at different gas pressures ( $p = 20, 30, \text{ and } 40 \text{ bar}$ ) and gas temperatures of 550 and 650 K. The initial droplet diameter was varied between 640 and 820  $\mu\text{m}$ . Their experimental results agreed well with theoretical predictions.

Other investigators have published results on multi-component fuel systems. Mikami et al (1997) studied the pressure effects in droplet combustion of miscible binary fuels composed of  $\text{C}_7\text{-C}_{16}$  hydrocarbons. They concluded that the burning lifetime shows a minimum value at pressures well in excess of the critical pressure of either fuel. Mikami et al (1998) also studied the interactive effects in two droplet combustion of miscible binary fuels at high pressure. They concluded that under subcritical conditions the quasi-steady region around a drop decreases with increasing pressure, and that the radius of the sphere decreases with increasing pressure. This suggests a reduction in the interaction coefficient with increasing pressure. This conclusion was substantiated by Harstad and Bellan (1998).

To facilitate studies under supercritical conditions, liquid fuel jets have been used. Chehroudi et al (1999) studied the initial growth and visual characteristics of a round jet in a sub- to supercritical environment of relevance to rocket, gas turbine, and diesel engines. They found that at low subcritical chamber pressures, the jets exhibit surface irregularities that amplify downstream. As the chamber pressure was raised, transition into a full atomization regime is inhibited at the critical pressure where both surface tension and heat of vaporization are sufficiently reduced. They concluded that the jet exhibits a dual nature: a liquid-jet and a gas-jet like faces depending on the values of

surface tension and heat of vaporization. Mayer et al (1996 and 1998) also examined the differences between subcritical and supercritical jet behavior.

Studies on transcritical cryogenic fluid jets by Woodward et al. (1994, 1996) yielded rich observations on the profound influence of transcritical effects on jet structure, atomization, and mixing (Talley, 1997). Raman imaging was successfully employed for the first time to obtain images that can be used to simultaneously quantify the ambient concentration of the species of interest as well as the vaporizing concentration field of the transcritical jet (Woodward et al., 1996). Their research also demonstrated that changes of mixture properties by doping the major liquid fuel with a small quantity of another liquid could have significant effect on the transcritical process. Under subcritical conditions, fuel jets will break up into droplets. These droplets will evaporate and gasify. Combustion then takes place. However, results from Woodward et al. indicated that droplets might not be formed from jet breakup at supercritical pressure, even at subcritical temperature.

The majority, if not all of the previous droplet experimental studies, has been confined to nonconvective or naturally convective conditions, which is no where close to the real situation in applications. Under the nonconvective and naturally convective conditions, heat and mass transfer between the droplet and the environment are limited by diffusion. Because the enthalpy of vaporization decreases with increasing pressure and approaches zero at the critical state, a droplet exposed to sufficiently high temperatures under supercritical pressures can presumably have more heat transferred to the liquid phase than is consumed by evaporative cooling such that continuous heating to criticality is quite possible.

Canada et al. (1975) and Kadota et al. (1981) considered the influence of natural convection on the droplet burning characteristics in their work. In addition to quiescent ambient, the experiments on the effect of forced convection on evaporation and combustion characteristics of suspended droplet was conducted by Jeng et al. (1990), Litchford et al. (1992), and Parigger (1994). In their research, first observations of critical transition phenomena were obtained of supercritical droplet gasification with forced convection without interference to the optical probing associated with a flame (Parigger, 1994).

Experiments with suspended droplet under supercritical conditions are theoretically impossible due to the loss of the surface tension caused by the disappearance of a distinctive liquid/gas interface when approaching the critical point. As a result, there will be no force to have the droplet suspended any more. Sato (1993) studied suspended droplets of heptane in nitrogen and calculated the emission constant from the  $d^2$  plots at sub- and supercritical temperatures and pressures. The observations showed that the emission rate becomes maximum at a pressure exceeding that of the critical point, and that this point of maximum emission rate occurs at decreasing pressures with increasing ambient temperature. These findings were confirmed by observations of Chauveau et al (1993) and Morin et al (1999).

The major experimental difficulties encountered in droplet combustion experiments are:

- (i) maintaining a spherical droplet shape and movement, and
- (ii) the ease of formation and release of the droplet under high temperature and pressure conditions.

Kumagai *et al.* (1971) used an electrical spark to ignite a suspended droplet at ambient pressure; even so, the electric spark still had some effect on the droplet's shape. Litchford and his co-workers (1992) used a pre-heated gas jet that came from the bottom of the combustor to ignite the suspended droplet through convective heating. During the ignition process, the aerodynamic force of the hot flow deformed the droplet.

The droplet formation and release under high pressure and/or high temperature are also difficult problems for droplet study. Litchford *et al.* (1992) and Hara *et al.* (1990) used tiny tubes to transfer the liquid fuel to a metal/quartz wire frame that was used for the droplet suspension. Because of the surface tension between the tube and the liquid, the droplet was difficult to transfer to the suspension wire, and the transferred droplet size was difficult to control. Furthermore, after the droplet was transferred to the suspension wire, it was more difficult to release to a free droplet. Above all, the loss of physical properties such as surface tension and heat of vaporization when the critical point is approached makes experiments on droplet combustion extremely difficult at high temperatures and pressures.

There are some questions as to the probable mechanism for gasification upon reaching criticality. Some might argue that because of intense internal circulation the droplet should undergo flash supercritical transition. Others might argue that because of temperature and concentration gradients, the droplet interior should remain subcritical when the surface reaches criticality so that subsequent gasification occurs through finite-rate regression of a critical interface. Still others might argue that a droplet should be dynamically unstable and disintegrate as a result of vanishing surface tension during

critical heating. These questions and uncertainties need to be addressed by solid experimental results.

Computational modeling work on hydrocarbon drop evaporation and combustion has become a major discipline of droplet study and has generated data and understanding of gasification and related phenomena. There are three levels of modeling proposed in the literature for the investigation of droplet behavior in different ambient environments (Bellan, 2000; Oran, 1991). These environments are defined based on the thermodynamic properties of the working liquid (existing as a droplet) and the thermodynamic state of surrounding gases. The three levels of modeling include: 1) droplet staying in a stagnant environment, 2) droplet experiencing a natural convection; and 3) droplet undergoing a forced convection with chemical reaction. In principle, under nonconvective conditions, heat and mass transport are purely diffusive in nature and droplet dynamics are irrelevant. Under natural convection or very mild forced convection associated with a free falling droplet, however, the transport is only weakly affected by convective effects and dynamic forces are minimal.

One of the earliest numerical modeling works was by Shuen et al (1992) which revealed detailed information on a liquid drop combusting under sub- and supercritical conditions. According to them, as ambient pressure is raised gas solubility in the liquid phase increases causing lowered critical mixing temperature. They also showed that flame radiation can play an important role for large drops (1000 micron) and must be accounted for in modeling studies.

Tsukamoto and Niioka (1993) studied the numerical simulation of heptane fuel droplet evaporation and ignition under high temperature and high pressure. The primary

qualitative conclusion from their work is that small drop ignition is controlled by the chemical reaction, whereas large drop ignition is controlled by emission process. Another set of investigations exploring high pressure conditions is that of Jia and Gogos (1992, 1993) and Gogos and Soh (1999). Using a n-hexane model in air for pressures of 0.1-10 MPa and temperatures 500-1250 K, the simulations showed that in the low temperature regime (~500 K) the drop lifetime increased with pressure, reaching a maximum around 600 K, further showing little sensitivity to the pressure, and finally at 1250 K decreasing with pressure. The droplet regression rate was found to be the same as the gas phase velocity.

The only study of bicomponent drops in a stagnant one-component environment at high pressure is due to Stengele et al (1999 and 1996). Their initial model (1996) studied a heptane-dodecane mixture (which was assumed ideal) in nitrogen and found that the  $d^2$  variation becomes linear past an initial heat up time during which the drop's size increases at supercritical conditions. This finding supports the study by Harstard and Bellan (2000) on an all-pressure fluid drop model applied to a binary mixture of heptane in nitrogen.

The latest contribution to the study of droplet behavior in convective, supercritical conditions is due to Gogos and Soh (1999). Their model studied a droplet of hexane in nitrogen in both micro- and normal gravity conditions. The axisymmetric conservation equations were solved including real gas effects, mixture non-ideality, solubility effects and variable properties for the two-phases. Their droplet lifetime predictions and the normal gravity data of Matloz et al (1972) agreed at low and moderate pressures, but deteriorated as the pressure increased. This deviation could be due to the difference

between suspended drop configuration of the experiment and the free floating drop of the simulation, as well as to the neglect of the thermal diffusion which becomes pronounced at high pressures.

The lack of data on isolated burning droplets in convective environments epitomizes the corresponding lack of fundamental information regarding hydrocarbon reaction rates in supercritical environments, and the uncertainties in treating the momentum transfer between what is optically identified as a droplet and its surroundings. These factors have combined to make accurate formulations impossible and, thereby making this topic area more challenging. The study at CAU will help provide the much needed experimental data to be used for theoretical studies of droplets at supercritical conditions. Droplet behavior in supercritical conditions with a strong forced convection needs to be addressed. The majority of the recent publications on droplet dynamics belong to this category (Hsiao et al, 1995; Lafon et al, 1995; Yang et al, 1992; Shuen et al, 1992; Liang et al, 1988). The key motivation for researchers to work in supercritical droplet environments with a strong forced convection is to cope with practical applications in airbreathing combustion, liquid fuel rocket propulsion and internal combustion engines.

Practical liquid-propellants are usually delivered to combustion chambers at a high pressure as a spray of droplets initially at subcritical temperatures, which are then heated up and experience a thermodynamic state transition into the supercritical regime. Subsequently, the sharp distinction between the gas and the droplet disappears and the whole system behaves differently from the current experimental observations at low pressure environments.



Despite the large amount of effort in the droplet combustion research area, transcritical droplet processes are still poorly understood due to the complexity of the processes involved and the poor understanding of thermodynamic and transport properties of mixtures under transcritical conditions. Significant difficulties in conducting transcritical experiments have also contributed to the lack of experimental data in this area. Thus, the previous research efforts have resulted in some inconclusive statements and certain plausible explanations, which provide a somewhat rational basis for anticipating the influence on and interaction with spray combustion dynamics in practical devices. Considerable uncertainties still remain. These include the fundamental basis on which any theoretical formulations are established. In order to develop a rational theoretical model, a comprehensive understanding of the physics of the vaporization process of the droplet is necessary.

In summary, it is believed that the actual path taken by the fuel droplets, based on the literature, should depend on the severity of the conditions to which the droplets are exposed. The direct observation of supercritical droplet behavior, particularly under different environments, is essential to resolving unanswered questions and enhancing the understanding of spray and droplet combustion. To seek conclusive evidence regarding the behavior of single component droplets in both inert and oxidizing atmospheres and at different sub- and supercritical environments through simplified and systematic experiments, is of urgent importance and is the major task of the research efforts in this area at Clark Atlanta University (CAU). Such experimental efforts will provide the much needed information for data validation of modeling results such as those being generated by researchers at the Jet Propulsion Laboratory (Bellan, 2000).

Three technical conference papers were published based on this research [4, 57, 58]. This is the final technical report of the contract period from March 1, 1998 to June 30, 2001. Journal publications are currently being prepared.

## **PROJECT TASKS AND ACCOMPLISHMENTS**

This project was scheduled into three phases. Phase I covered the project initiation phase and included:

1. Updating of the literature,
2. Modification and installation of experimental setup,
3. System integration and synchronization,
4. Data image processing software development and application,
5. System shake-down and detail experiments with hexane and other hydrocarbons, and
6. Joint experiments with the Air Force Research Laboratory (AFRL).

Phase II of the study involved experiments that focused on characterization of the interaction between the droplet and the ambient environment and the droplet behavior during the processes of heating and evaporation. Phase III focused on vaporization and combustion experiments at sub and supercritical environments. To date, all of the planned activities as described in the project milestone chart were completed.

This project provided very revealing qualitative information on the gasification and combustion behavior in air and valuable quantitative data on droplet size variation with temperature, pressure and molecular weight. Clark Atlanta University intends to

continue to be a major player in this field. What is needed most at the end of the current project is a simplified, systematic study that will:

- isolate some of the influences (e.g. ignition and combustion chemistry) that complicate the gasification behavior at sub- and supercritical conditions,
- be coordinated with modeling efforts to provide relevant data (e.g. initial and boundary conditions) for data validation.

Among the desired data that will be acquired in the future simplified and systematic studies are:

- Quantitative data on single fuel drops in inert and oxidizing atmospheres;
- Data for single component fuels under forced convection conditions;
- Data on composition, density and temperature profiles;
- Data on velocity distribution in the phases within the gasification/combustion chamber; and
- Detailed understanding, analysis and modeling of the results for droplets.

## EXPERIMENTAL

### *Materials*

Analytical grade hexane, heptane, decane and tetradecane were used for the molecular weight, temperature and pressure variation experiments. Some physical data of the hydrocarbons are listed in Table 1.

**Table 1.** Physical properties of selected hydrocarbons

Compound	Molecular Weight	Critical Temp (°C) [15]	Critical Pressure [15]	
			(MPa)	(Psi)
Hexane (C <sub>6</sub> H <sub>14</sub> )	86	507.7	3.010	436.6
Heptane (C <sub>7</sub> H <sub>16</sub> )	100	540.3	2.756	399.7
Decane (C <sub>10</sub> H <sub>22</sub> )	142	617.7	2.104	305.2
Tetradecane (C <sub>14</sub> H <sub>30</sub> )	198	693.0	1.62	235.0

### *Experimental Set-up*

The droplet combustion experiment hardware designed and built by the project team and shown schematically in Figure 1, consists of a liquid hydrocarbon fuel supply system, a gas pressure-control system, a droplet formation and release system, a cylindrical high pressure and temperature chamber, a Vulcan Electric 304 stainless steel heater, an electrical ignition system, and a high-speed Kodak EktaPro CCD (charged-coupled detector) video system.

### *Liquid Fuel Supply System*

The liquid fuel supply system also designed and built by the project team and shown in Figure 2 consists of a liquid-fuel reservoir, and an Eldex Laboratory Model B-100-S high-pressure liquid pump. The pump can operate up to 1500-psi pressure and provides flow rates from 0 to 150 mL/min. The liquid fuel was injected into the combustion chamber through a 1/16" 304 stainless steel tubing. A needle valve and the high-pressure pump adjusted the rate of flow of liquid into the combustion chamber.

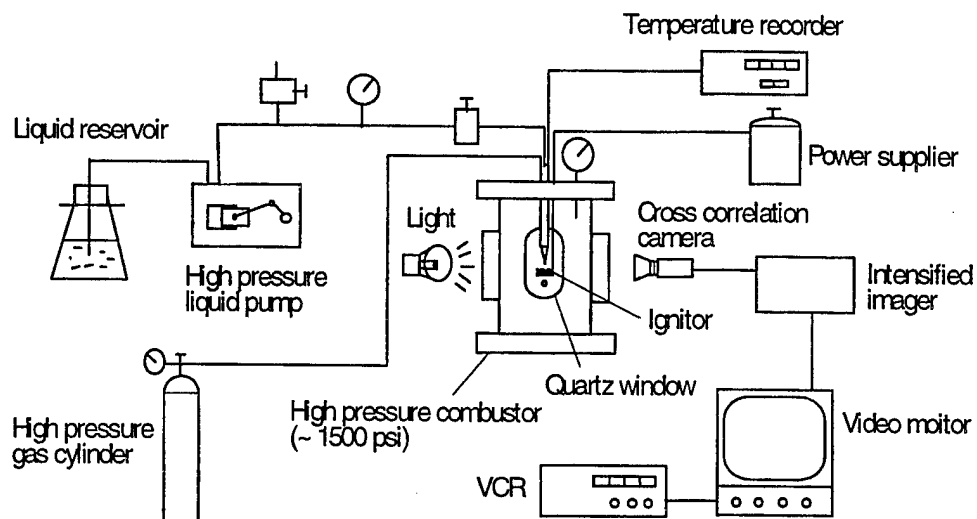


Fig. 1. Schematic of the experimental setup

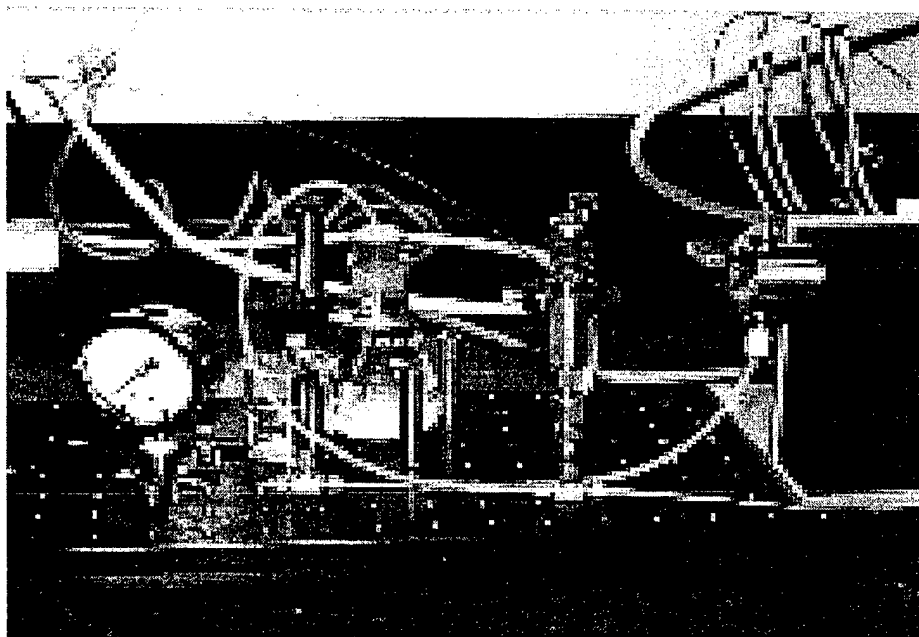


Figure 2. The Liquid Fuel Supply system

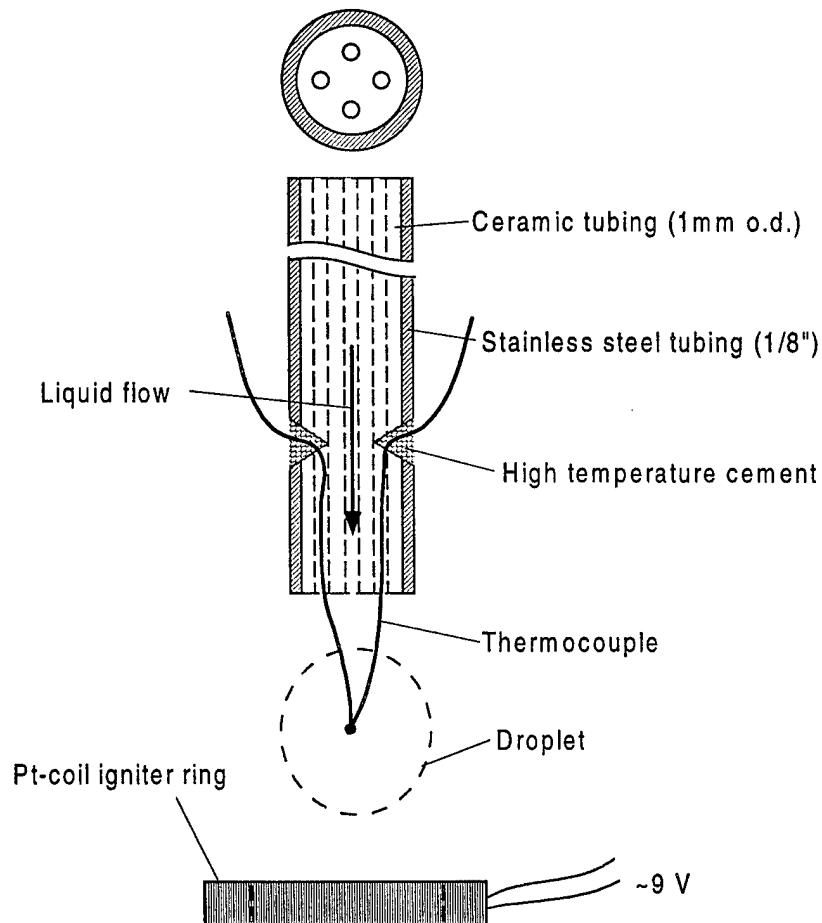


Figure 3. Droplet formation system and igniter

#### *Droplet Formation and Release System*

The droplet formation and deployment system, designed and constructed for this study, consisted of a  $200 \times 1$ -mm-o.d. ceramic tube, which had four 0.2-mm-i.d. holes in its axial direction and a thermocouple. At the position of 30 mm from one end, the wall of the ceramic tube was cut to expose two of the four holes (Figure 3). The thermocouple's two wires were inserted into the two holes of the ceramic tube and introduced out from the two cuts on the tube wall, where the tip of the thermocouple was

left about 10 mm from the end of the ceramic tube. The ceramic tube was then inserted into a 1/8" stainless steel tube that also had two cuts on its wall through which the thermocouple wire passed. The cuts were sealed by high temperature cement. The stainless steel tube was vertically mounted on top of the combustor, while the ceramic tube was connected to 1/16 stainless steel tubing of the liquid feeding system by a stainless steel union.

The pressurized liquid fuel was introduced to the other two unused holes of the ceramic tube. The fuel exited from the other end and attached to the thermocouple wires to form a 1.5-mm-dia droplet at the tip of the thermocouple. The rate of droplet formation was controlled by the liquid flow rate.

### *Electrical Igniter*

The electrical ignition system of the study consisted of a filament and power supply. The filament was made from a coiled 0.8-mm diameter nickel-chromium wire, and was wound into a 4-mm-i.d. ring and mounted 20-mm below the tip of the thermocouple. The igniter ring was carefully aligned so that the free droplet passed through the ring without touching it (Fig. 3). The temperature of the ignition filament was controlled from ambient to 1400 °C by adjusting the voltage of the variable autotransformer. About 800 °C was high enough to ignite the hexane free droplet in this study, with the power supply voltage at about 9 V.a.c.

### *High Pressure Chamber*

The chamber used was cylindrical and had a test section of 450 (high)  $\times$  95 (dia.) mm, with four quartz windows of 90 (high)  $\times$  38 (wide). The high pressure and high temperature reaction chamber was capable of operation in the temperature range of 25 to 600 °C and pressure range of 14.7 – 12,000 psi. Such a reaction system enabled experiments to be conducted that covered both sub- and supercritical temperatures and

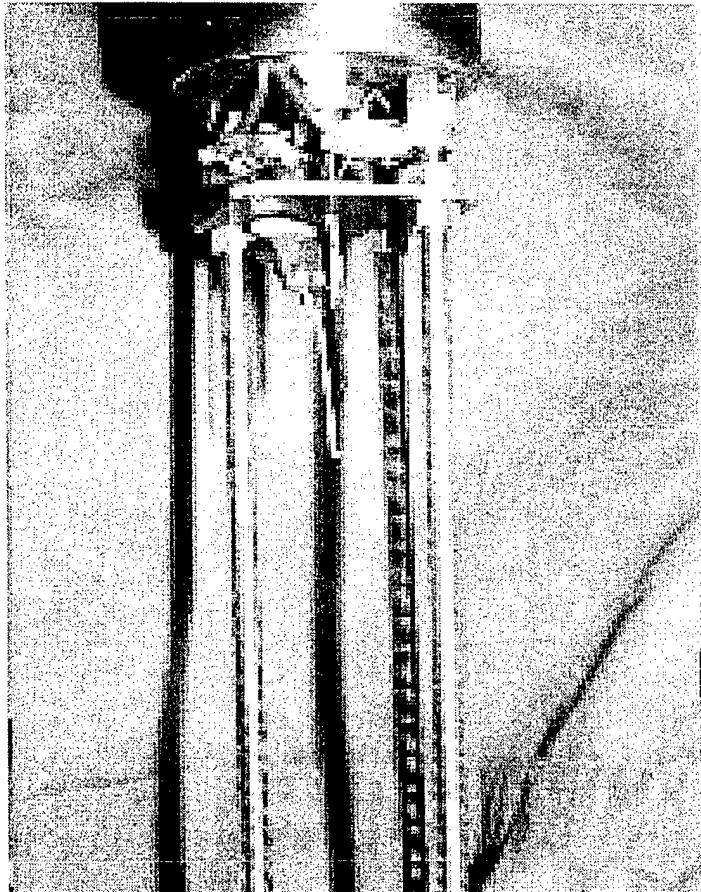


Figure 4. Picture showing the droplet formation and release system with igniter ring



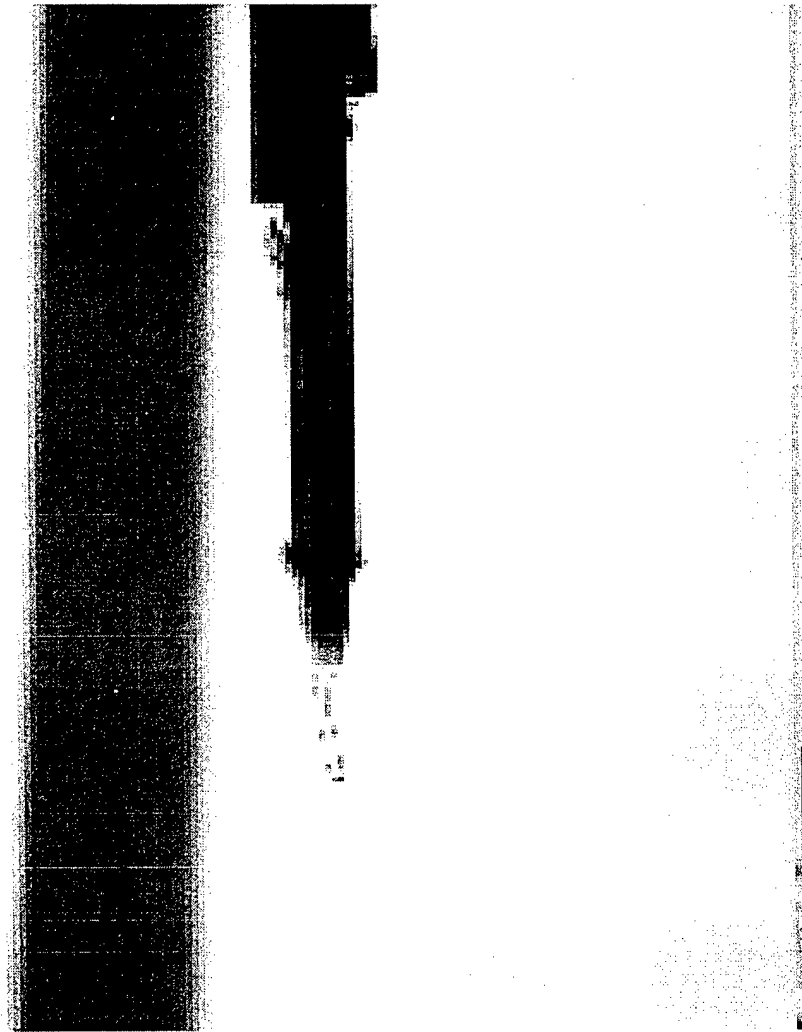


Figure 5. Picture showing tip of thermocouple on which the droplet is suspended.

pressures of the fuels ( $C_6$  to  $C_{14}$ ) studied. The use of quartz windows allowed for visualization measurements. To provide a convective heating environment for the gasification and combustion studies, a different chamber was constructed as shown in Figure 6. The chamber contained a Caloritech Inc. gas burner, a ball valve, and a honeycomb flow straightener.

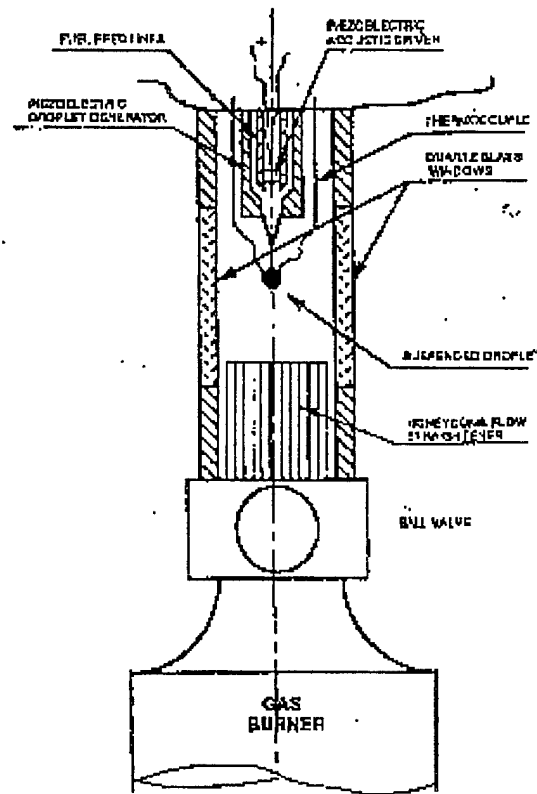


Figure 6. Combustion chamber for convective environment droplet study

### *Electrical Heater*

The electrical heater, shown in Figure 7, is a Vulcan Electric 304 stainless steel sheath with high temperature leads suitable up to 650 °C. The heater has an overall length of 165.1 mm ( 6.5 in ) and has a mounting dimension of 190.5 mm ( 5.5 in ).

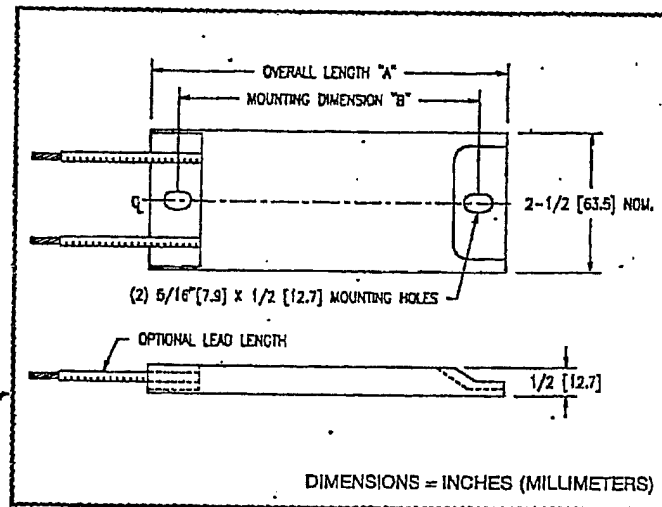
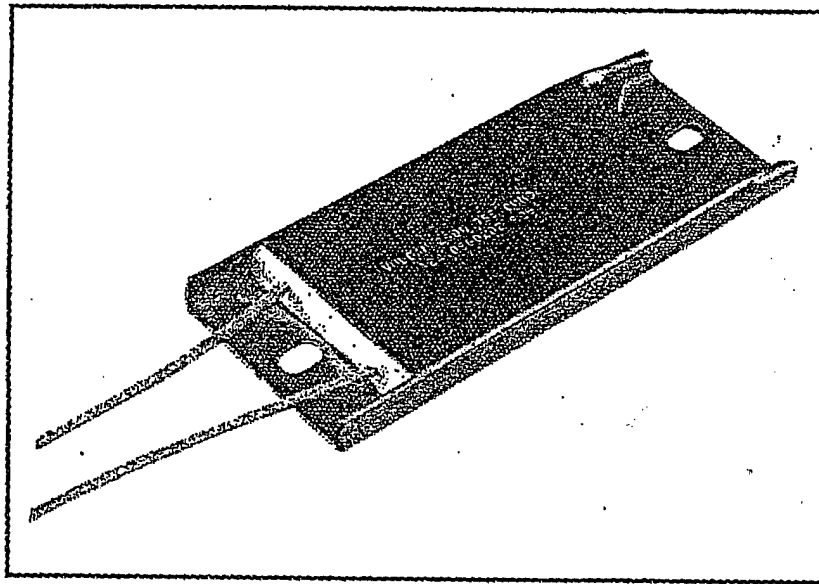


Figure 7. A Strip Heater made of 304 Stainless Steel Sheath with High Temperature Leads

Two 450-Watt strip heaters, each one with a watt density of  $3.1 \text{ W/cm}^2$  ( $20 \text{ W/in}^2$ ), were connected in series and installed on the base of the combustion chamber.

Temperature control was achieved by using a variable autotransformer, supplied by Staco Energy Products Company, with an output voltage of 0-120/140V, a current of 10 A and frequency of 50/60 Hz.

### *Optical Measurement System*

The optical measuring system in Figure 8 is composed of a Kodak EktaPro Hi-Spec

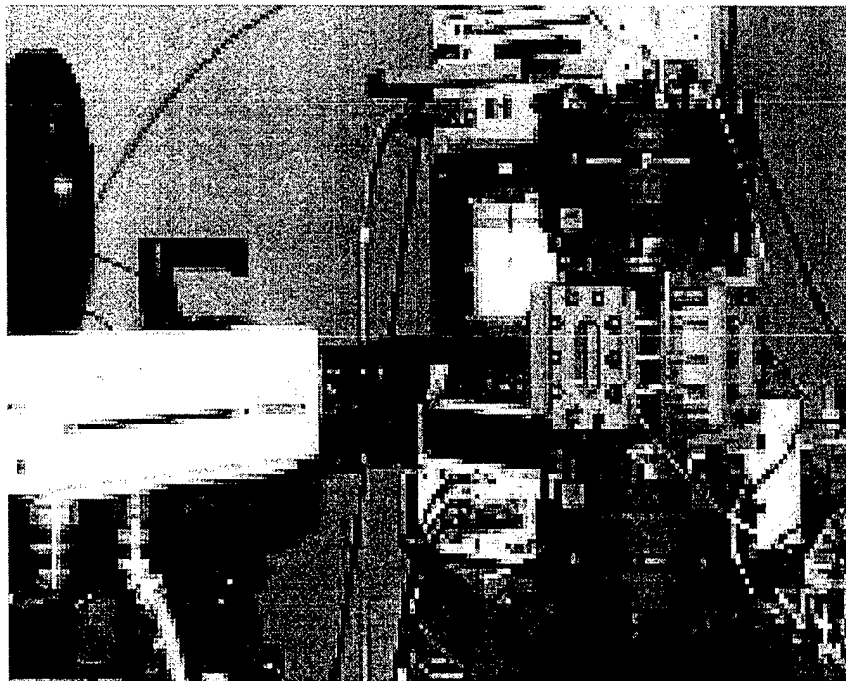


Figure 8. Experimental setup showing the combustion chamber and the optical measurement system.

Intensified Imager, which consists of an industry standard 2-5/8" x 4-3/4" dovetail adapter plate and a Nikon micro lens. The imaging system also includes an EktaPro

Intensified Imager Controller and an EktaPro Hi-Spec Processor. All of the images were continuously taken at a speed of 1000 frame pictures per second (fps/s) with an exposure time of  $\sim 30 \mu\text{s}$ .

### *Experimental Procedure*

The experiments were conducted using the following procedures: (1) Because of the extensive heat loss encountered in high temperature studies, the combustion chamber was insulated with a Miraflex multi-project thermal and noise control insulation material. The heater was heated to the desired temperature using a variable autotransformer. (2) the combustion chamber was pressurized to the desired value with compressed air, and the liquid pump was started which pressurized the liquid fuels. (3) For the gasification rate measurements, a suspended droplet was used. The inlet needle valve on the liquid feeding line was opened first and the flow rate carefully adjusted until a droplet hung on the tip of the thermocouple. Then, the needle valve was quickly closed. The droplet image was then recorded using the video system. (4) For the combustion tests, a free droplet was used. The liquid flow rate was carefully increased to release the droplet after forming at the tip of the thermocouple. While the free droplet was falling through the igniter's filament ring, it was ignited. The burning droplet continuously fell downward or floated back upward depending on the system conditions. The whole process was recorded by the high-speed video system and analyzed.

The following system temperatures and pressures ranging from the subcritical regime to the supercritical regime were studied: (1) low p, low T, (2) low p, high T, (3) high p, low T, (4) high p, high T.

## RESULTS AND DISCUSSION

### *A. Droplet Gasification*

Because of its small size and compact design, the system developed at Clark Atlanta University can be used for both normal and microgravity tests in a ground station or in a microgravity environment. Figure 9 shows that the square of the diameter of the hexane droplet vs. time is a straight line in most parts of the gasification period. This linearity can be explained by surface effect. Because the gasification occurs on the surface of a droplet, at a constant temperature the gasification rate is proportional to the surface area of a droplet. The surface area of a droplet, assumed as a sphere, is a function of the square of its diameter. However, Fig. 9 indicates that after the diameter squared diminished to about  $0.50 \text{ mm}^2$  ( $\sim 0.8 \text{ mm}$  in diameter) at 200 and 800 psi system pressure, the hexane gasification rate increased by a factor of about three. This increase may be caused by the following effects. As the droplet diameter decreases, its curvature increases, and its surface tension (energy) increases. As a result, the escaping tendency of the molecules at the droplet surface becomes stronger, and its gasification rate per unit surface area increased. In addition, the gasification rate was measured from a suspended droplet. Because of the effects of gravity and the thermocouple wires, the droplet was found not to be a perfect sphere but with some deformation. As the droplet size decreased, the deformation was observed to increase in this study. The combination of these two effects is believed to be responsible for the change in gasification rate of the droplet as its size decreased.

Figure 9 further shows that the hexane gasification rate at the 500 psi system pressure was more than double those at 200 and 800 psi. This increase is thought to be

the effect of supercritical conditions. The critical pressure of hexane is 436.6 psi (3.01 MPa). Under system pressures close to or above the hexane critical pressure and ambient temperatures less than hexane's critical temperature, the molecules become more excited, and result in a faster decrease in the size of the denser fluid (original droplet) than that under sub-critical pressure conditions. After passing the critical point, as the pressure further increased, the molecular excitement did not change much, but the increased pressure decreased the fluid molecule's escaping tendency. The gasification rate of the suspended droplets of heptane and decane, like the hexane, reached their maximum value at their corresponding critical pressures.

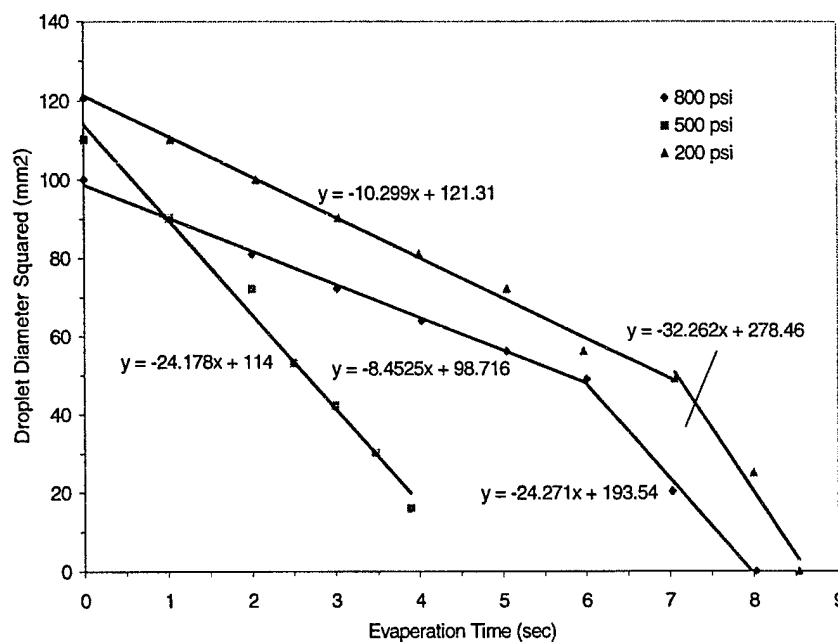


Fig. 9 The changes of the surface areas (diameter square) of hexane droplets with gasification time at different pressures.

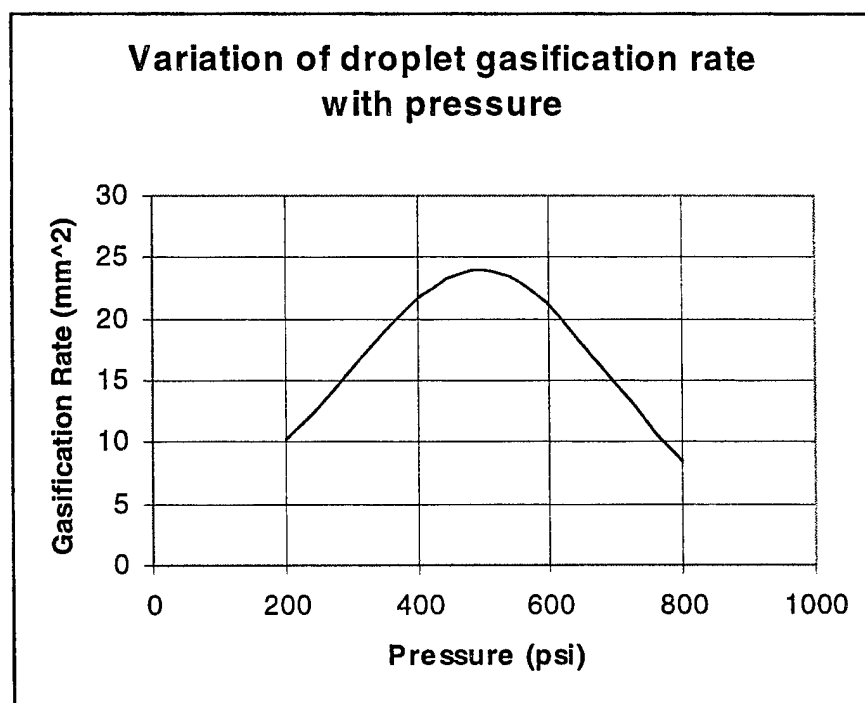


Figure 10. The effect of system pressure on the gasification rate of hexane at room temperature.

Figure 10 shows a plot of the surface area, measured as diameter squared, with pressure. The gasification rate was found to increase with increase in pressure and temperature at subcritical conditions to a maximum at the critical pressure of hexane and then decreased at supercritical pressures.

Figures 11 and 12 show the variation of hexane and heptane droplet diameters with gasification time at different temperatures and ambient pressure. The results indicate that both n-heptane and n-hexane droplet exhibit classical  $d^2$ -law gasification behavior at subcritical temperatures, as the gasification rate is nearly constant over most



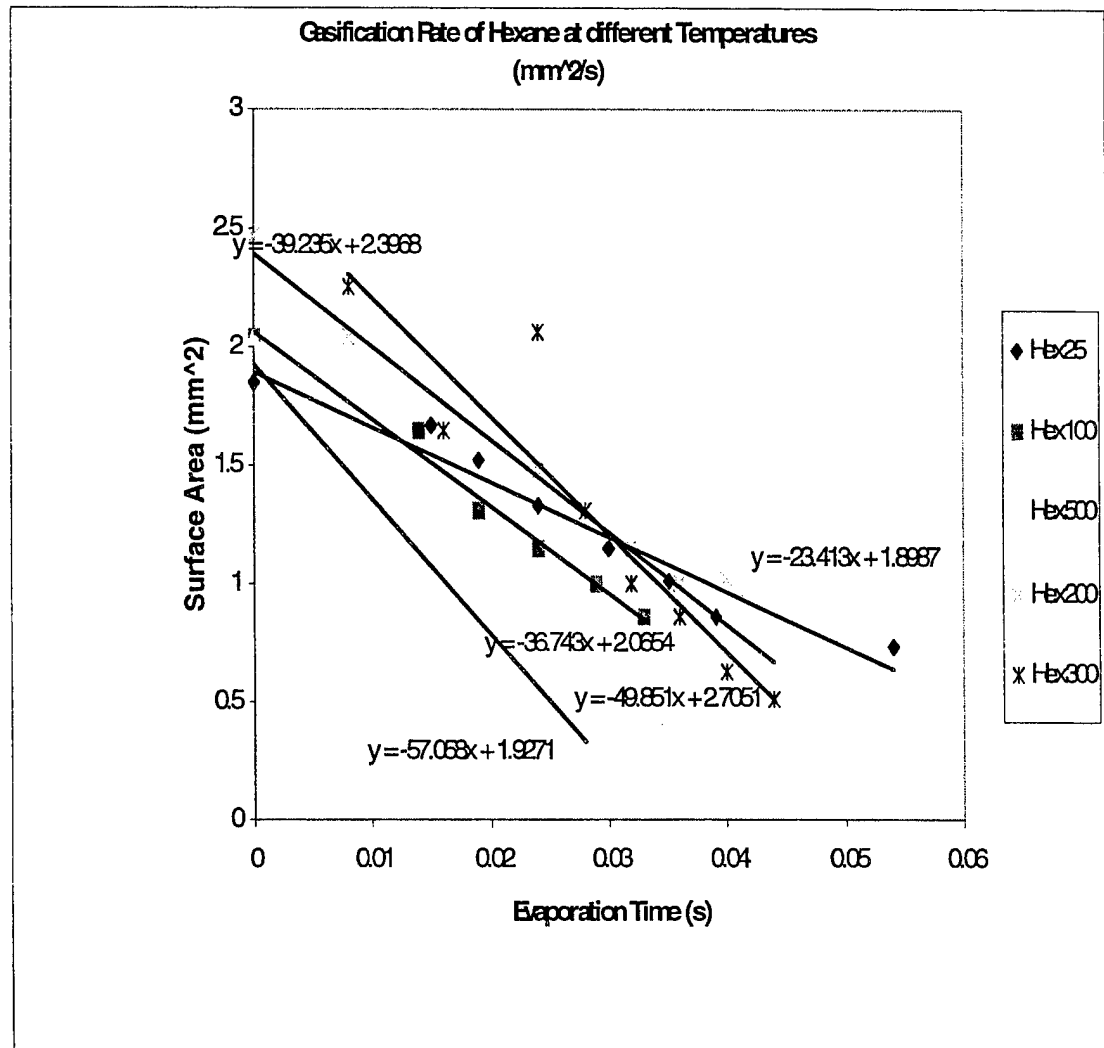


Figure 11. The changes in surface area of hexane droplet (measured as diameter squared) with gasification time at different temperatures and ambient pressure.

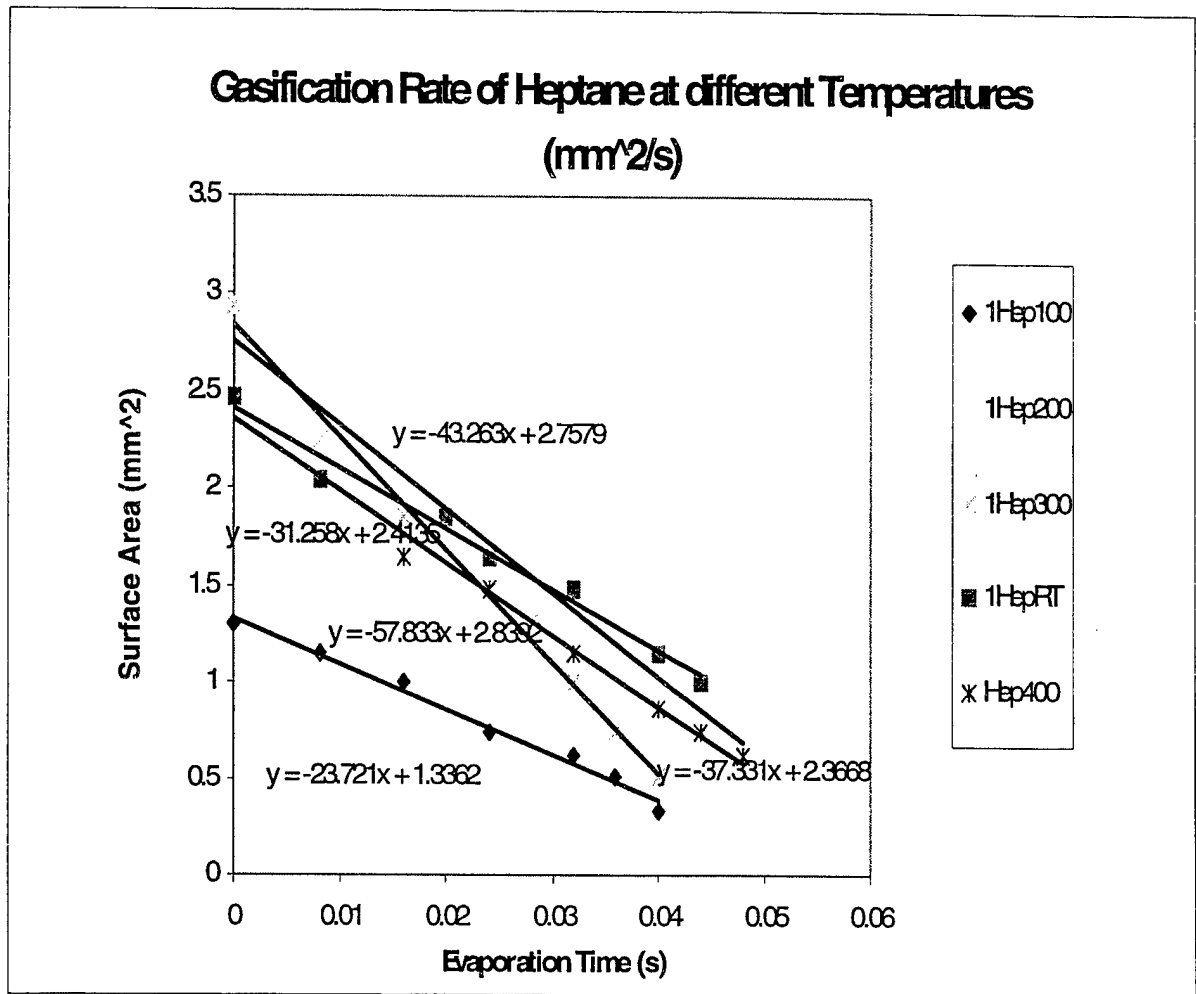


Figure 12. The changes in surface area of heptane droplet (measured as diameter squared) with gasification time at different temperatures and ambient pressure.

parts of the gasification period. This linearity can be explained by surface effect. The increasing trend is due to the fact that molecular motion is enhanced at higher temperatures. The escaping tendency of the molecules at the droplet surface becomes stronger, and therefore, the gasification rate increased.

Figure 13 shows a plot of the variation of the gasification rates of hexane and heptane droplet with temperature. The gasification rate was found to increase non-

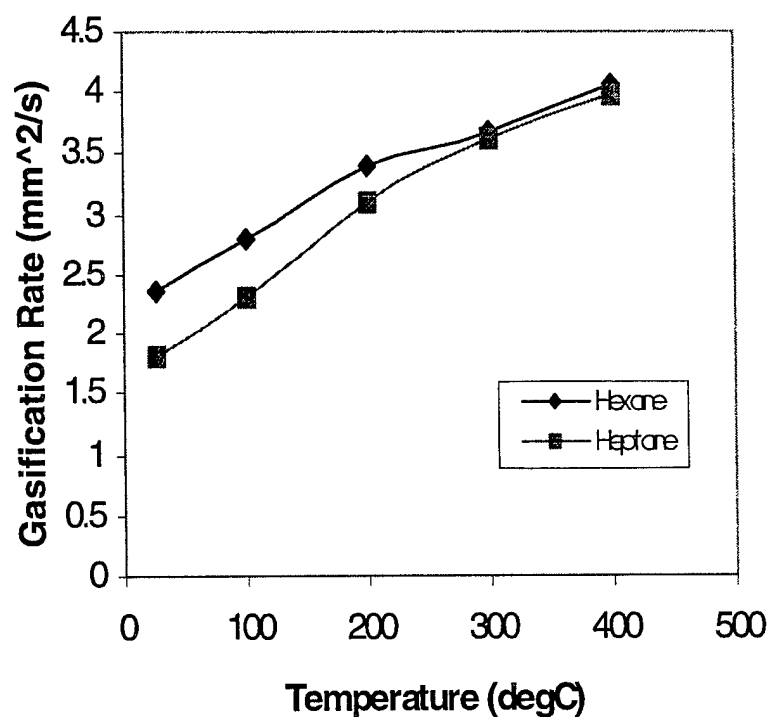


Figure 13. Variation of gasification rate with temperature for hexane and heptane.

linearly with increase in temperature at temperatures below the critical temperature for both fuels. However, the rate was higher for hexane. This is to be expected since the molecular weight of hexane is lower than that of heptane.

The hexane droplet behavior around the critical temperature was studied by suspending a droplet at the tip of the thermocouple and increasing its temperature from 400 to 500 °C. It was observed that as the droplet temperature increased to ~500 °C, (critical temperature for hexane is 507.7 °C) the droplet became unstable and disintegrated. This is attributed to the vanishing surface tension at the critical temperature.

The dependence of the fuel molecular weight on droplet gasification was also studied for fuels ranging from hexane to tetradecane. Figure 14 shows the variation of gasification rate with fuel molecular weight. At ambient temperature, the results indicate that the gasification rate of a hydrocarbon droplet proportionally decreases with the molecular weight of the hydrocarbon under a fixed temperature and pressure condition, but it reached its maximum value at the critical pressure of each compound. This decrease occurs because as the molecular weight of a hydrocarbon increases, its size increases and its vapor pressure and activity decrease.

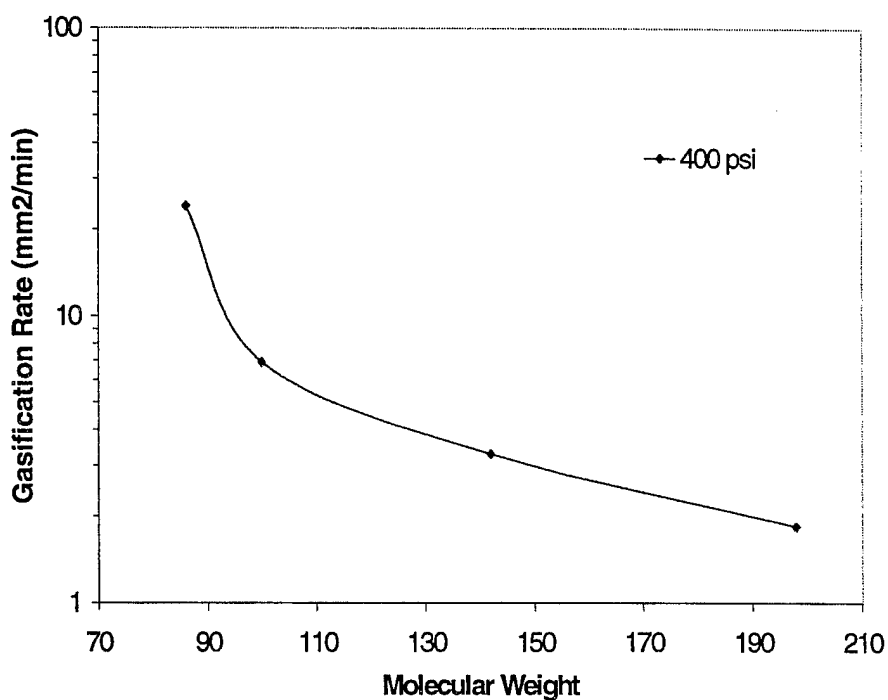


Fig. 14. Variation of molecular weight of hydrocarbon fuels with gasification rate

The combined effect of temperature and pressure on the gasification and combustion characteristics was studied. Figure 15 shows the variation of the gasification rate of hexane with pressure and temperature. Results show that the gasification rate increases with increase in pressure and temperature up to the critical pressure. Above the critical pressure the gasification rate decreases.

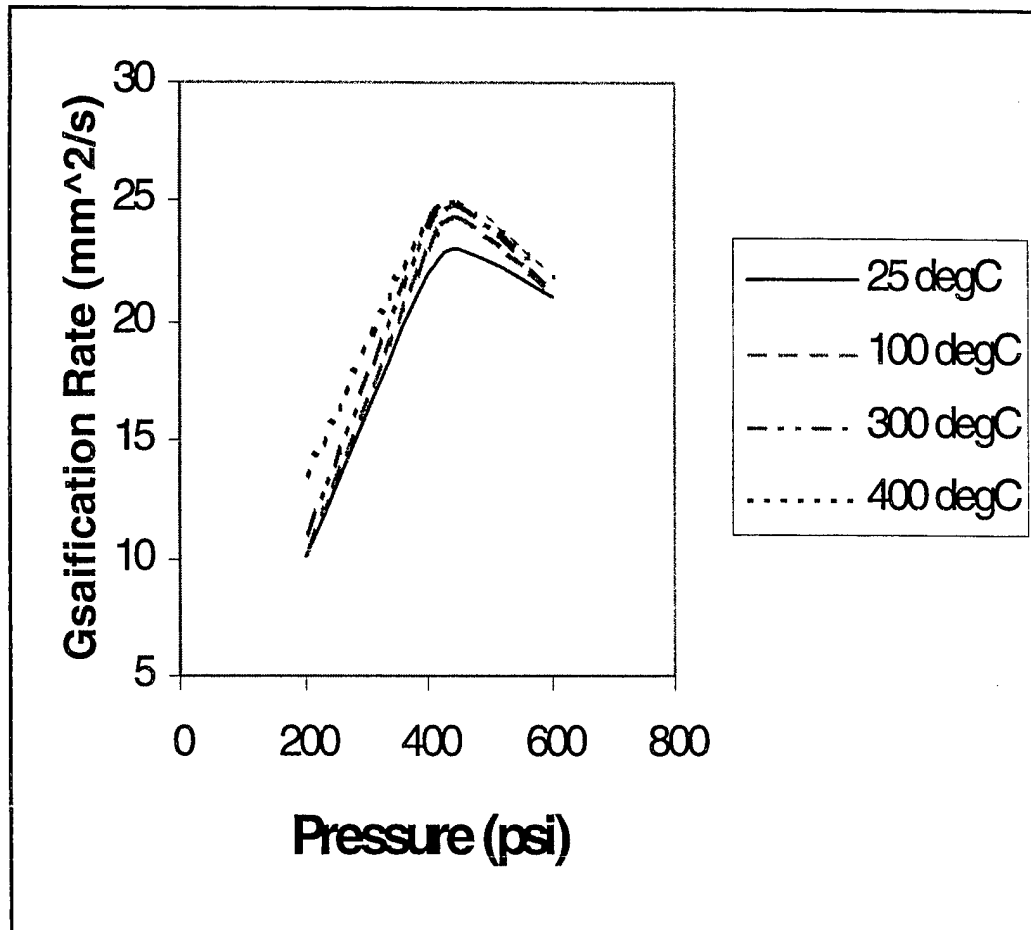


Figure 15. The variation of the gasification rate of hexane with pressure and temperature.

### *B. Free Falling Droplet Tests*

When a droplet was released from the thermocouple, it started to fall. The whole process was recorded by the high-speed video system at a recording speed of 1000 frames per second with an exposure time of about 30 microseconds. From replays of the recorded images at a much slower speed (e.g. 1 – 30 frames/sec), the shape and speed of

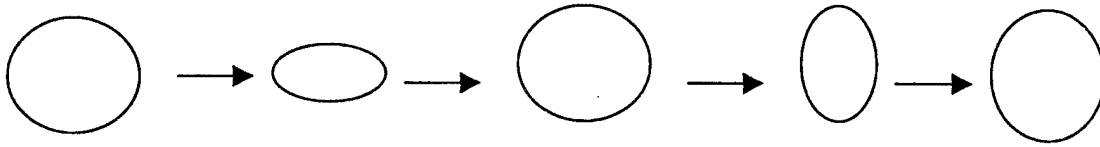


Figure 15. Oscillatory deformation of a free drop droplet of hexane

the falling droplet were observed and measured. The oscillatory deformation of the droplet as it fell is illustrated in Figure 15. It was found that during falling, the shape of a droplet oscillated in the vertical direction. The magnitude of the oscillation changed with the system temperature and pressure. The deformation of the droplet was enhanced as the temperature and pressure increased.

Figure 16 shows the variation of the droplet acceleration with temperature at constant pressure. The effective acceleration of a free-falling droplet of hexane was found to increase linearly with increase in temperature at subcritical temperatures when the system pressure was held constant at one atmosphere. The increase is due to the fact that as the temperature of the system is increased, the gas density in the combustion chamber decreases, allowing the droplet to fall rapidly.

The variation of the resistance force with temperature is shown in Figure 17. The resistance force,  $F$ , was calculated according to  $F = \frac{V}{\rho} \alpha$ , where  $V$  and  $\rho$  are the volume and density of the droplet respectively and  $\alpha$  is the droplet acceleration. At subcritical temperatures, the resistance force decreased with increase in system temperature at ambient pressure.

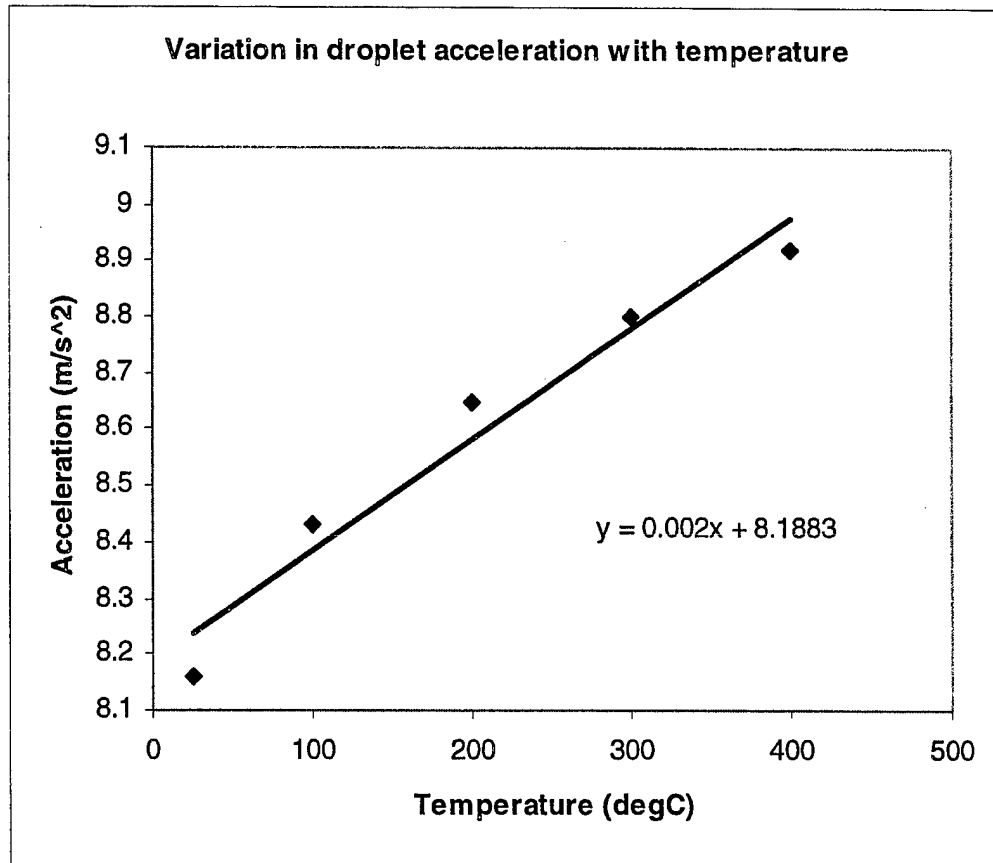


Figure 16. The effect of temperature variation on hexane droplet acceleration.

Figure 18 shows that the effective acceleration of a hexane free droplet,  $\alpha$ , is inversely proportionally to the increase of the system pressure from ambient (14.7) to 1200 psi. This proportionality is due to buoyancy. For a free falling droplet, gravity is the driving force, and buoyancy the resistance force. As the system pressure increased,



the resistance force increased, while gravity remained constant. This balance is shown in Figure 18. Figure 18 indicates that as the system pressure approached the droplet's critical pressure ( at 436.6 psi for hexane ) no obvious effect on the hexane

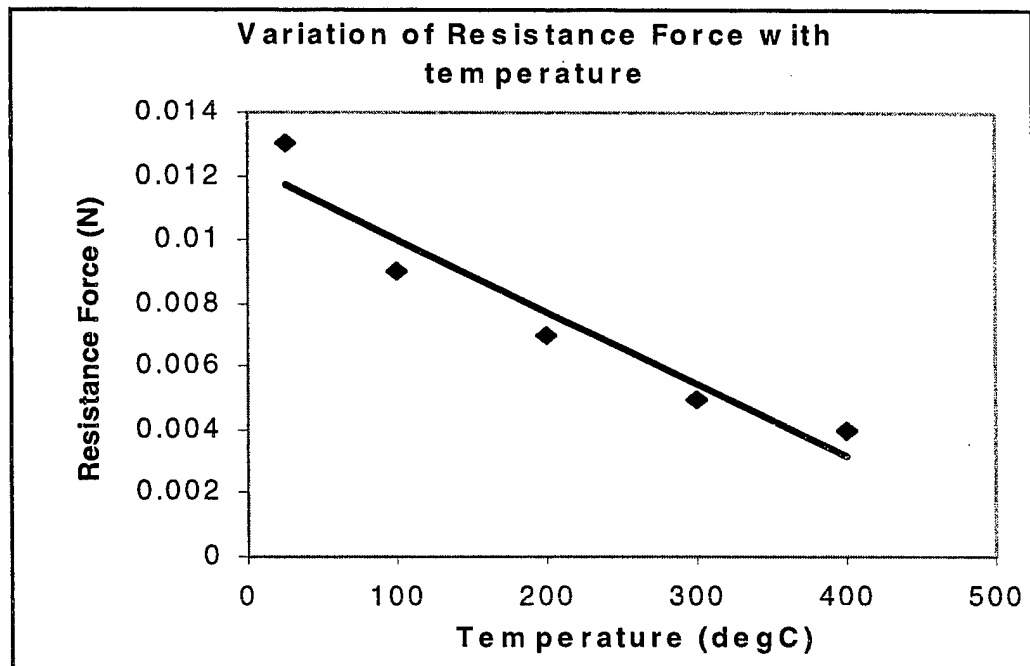


Figure 17. The effect of temperature on the resistance force on hexane droplet at ambient pressure.

falling acceleration was observed. This is because the hexane partial pressure was below its critical pressure under the experimental conditions.

At system pressures close to the decane critical pressure (300 psi), the non-combusting free-drop droplet of decane approached its maximum falling velocity of 4.23 m/s. When the pressure moved from the critical pressure to either lower or higher pressure, the falling velocity of the droplet proportionally decreased. As the system

pressure decreased to 200 psi and 14.7 psi (ambient), the falling rate of the droplet decreased to 3.97 and 2.15 m/s, respectively. As the system pressure increased to 400 and 600 psi, the falling rate decreased to 4.09 and 3.28 m/s, respectively.

### *Free Falling Droplet Combustion Tests*

From the recorded video images, it was observed that the system pressure had significant effects on the combustion of a free falling droplet. As the system pressure increased, the falling speed of a droplet proportionally decreased. When the chamber pressure was increased to a certain value (~200 psi), after falling below the igniter ring for a distance, the burning droplet started to float upwards. As the chamber pressure increased, the distance that the burning droplet fell below the igniter ring before floating

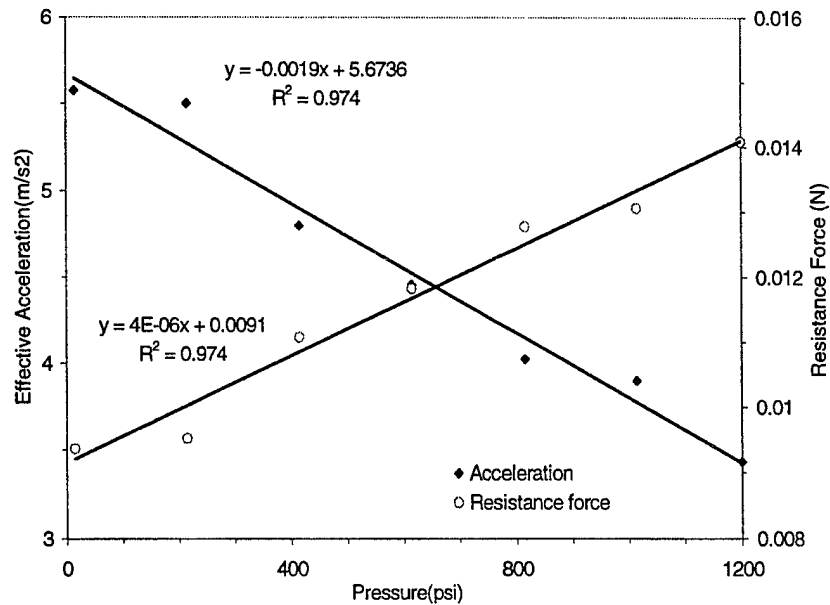


Figure 18. The effect of the system pressure on the effective acceleration and resistance force of a free falling hexane droplet.

upwards decreased as shown in Figure 19. As the chamber pressure approached the droplet's critical pressure, the free burning droplet virtually stayed at its ignited position (Figure 19). This phenomenon can be explained by the effect of buoyancy. After a free droplet was ignited, the gas around the droplet was heated and became less dense. Differences in densities of cold (unheated) and hot (heated) gases resulted in a buoyancy force that helped to push the droplet upwards. As the system pressure increased, the gas density in the combustion chamber increased, and the differences in densities between the hydrocarbon droplet and gas decreased.

Figure 20 indicates that the system pressure also has some effect on the flame life of a droplet, which is defined as the combustion time of the droplet. At about 600 psi (near the critical pressure), the hexane droplet had the longest flame life. As the pressure moved from that value, the droplet's flame life decreased. At the ambient pressure, a droplet was measured to have a longer flame life than that at 200 psi, which was probably caused by the low oxygen potential at ambient pressure because air was used as the combustion gas.

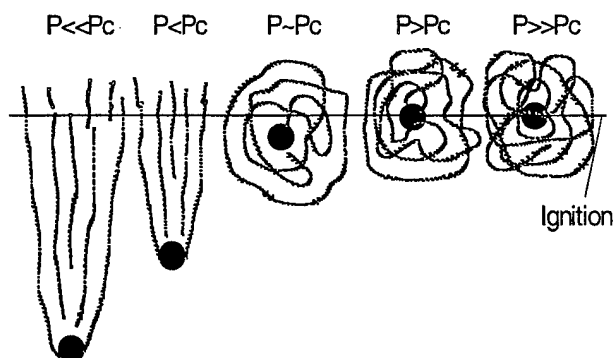


Fig. 19. The effect of the system pressure on hexane free-falling burning droplet motion.

As the molecular weight of the hydrocarbons and the system pressure increased, the ignition of the free-drop droplets became more difficult, the combustion rates of the droplets decreased, and the flames of the combusting free-drop droplets became less stable. As shown in Table 2, all of the free-drop droplets of hexane under the system pressure from ambient (14.7 psi) to 1200 psi were ignited. The power voltage needed for the ignition proportionally increased from 7.0 to 9.0 V as the system pressure increased from ambient to 1200 psi. The increase of the power voltage required for the ignition may be caused by two factors. One factor is the decrease of the igniter temperature. As the system pressure increased, the thermal conductivity of the gas increased, so the igniter's temperature decreased. This temperature decrease was observed from the color change of the heated igniter during the experiments. The other factor is the decrease of the droplet vapor pressure with the increase of the system pressure. For liquid hydrocarbons, since only their vapor phase can be ignited to form a stable flame in the air, the decreased vapor pressure makes the ignition and combustion more difficult.

Table 2 shows that all of the free-drop droplets of heptane under the system pressure from ambient to 1000 psi were ignited, at a higher power voltage than that required for hexane under the same system pressure. Also, it was observed that during the experiments the free-drop droplets of hexane were ignited earlier (relative to the position of the igniter) than those of heptane. This difference in time should also be caused by the vapor pressure difference of hexane and heptane. As the droplet fell, a concentrated vapor sphere surrounded and moved with it. When the droplet was heated

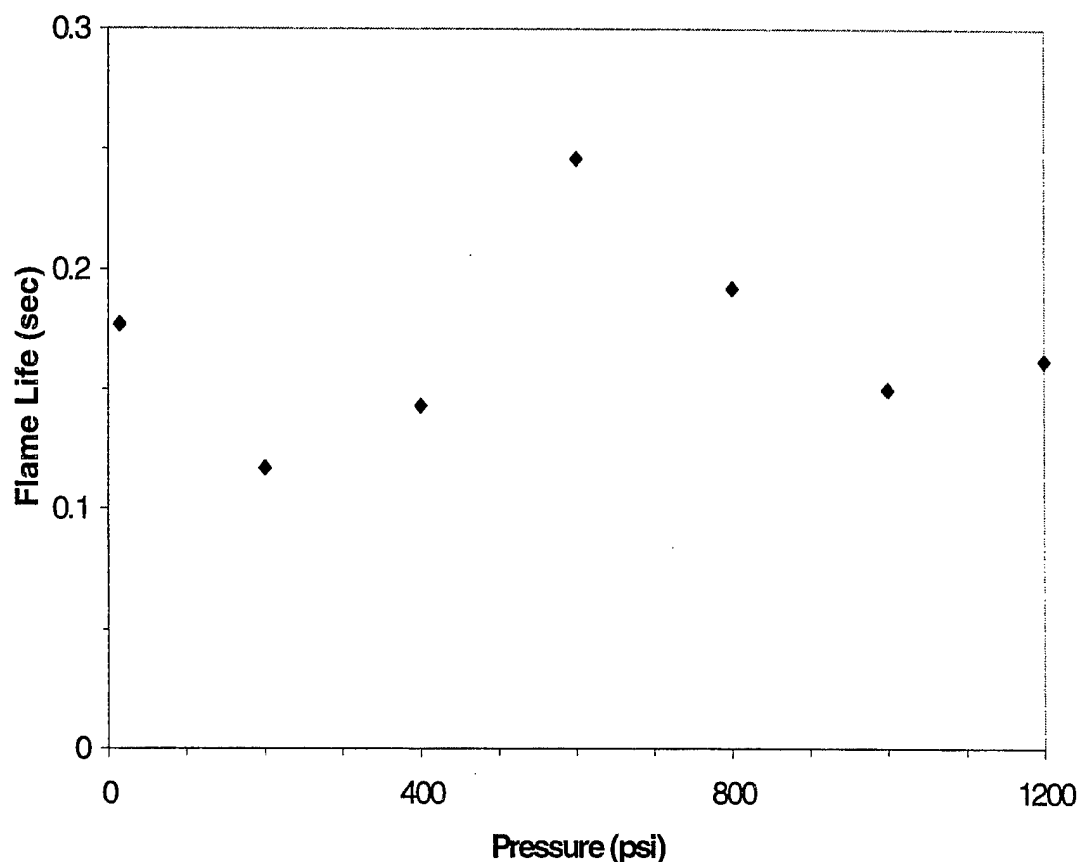


Figure 20. The effects of the system pressures on the flame life of a free falling droplet.

this vapor could be observed clearly from the high-speed video images. Hexane's higher vapor pressure led to a larger surrounding vapor sphere that reached the igniter earlier and was ignited earlier. The size of concentrated vapor sphere of a droplet was found to be proportionally decreased as the system pressure increased, but no large change was observed after the system pressure passed through the hydrocarbon's critical pressure.

**Table 2** The igniter power voltage needed for the ignition of the free-drop droplets of several hydrocarbons under the different compressed-air system pressures

System Pressure (psi)	Power Voltage Needed			
	Hexane	Heptane	Decane	Tetradecane
Ambient (14.7)	7.0	7.0	Not ignited*	7.0
200	7.0	9.0	Not ignited	10.0
240	-	-	-	10.2
300	7.5	-	Not ignited	10.2
400	8.0	10.0	Not ignited	10.4
600	9.0	10.0	Not ignited	11.0 **
800	9.0	10.3	Not ignited	11.0 **
1000	9.0	10.6	Not ignited	11.0 **
1200	9.0	-	-	-

\* The free-drop droplet could not be ignited at igniter power voltages up to 14 V.

\*\*The free-drop droplet was ignited at the igniter, but the flame extinguished after the droplet passed the igniter.

As the number of carbon atoms of a droplet hydrocarbon molecule increased to 14 (tetradecane), the free-drop droplets were ignited in all of the system pressure range from ambient to 1000 psi but with quite different behaviors. At the ambient pressure, the free-drop droplet of tetradecane was ignited at a power supply voltage of 7.0 V and combusted in a similar way to the droplets of hexane and heptane. As the system pressure increased to near and above the critical pressure of tetradecane ( $\geq 200$  psi), the ignition voltage increased to 10 V, and the combustion rate of the droplet decreased. When the system pressure was two or more times that of the critical pressure ( $\geq 600$  psi), the free-drop

droplet was ignited at 11 V, but the flame of the free-drop droplet existed only near the igniter. As soon as the droplet passed through the igniter, the flame extinguished.

The free-drop droplets of decane (C<sub>10</sub>) could not be ignited under any of the experimental conditions used, from ambient to 1000 psi of the system air (21% oxygen and 79% nitrogen) pressure and up to 14 igniter V (~1400 °C). When the free-drop droplets of decane passed through the igniter, some smoke formed at the droplet. As the system pressure increased, the smoke became more intense. Additional experiments are needed before one can explain the observed puzzling behavior of decane droplet combustion.

It was found that increasing the system's oxygen partial pressure led to enhanced ignition, combustion rate, and flame stability of the free-drop droplets.

## CONCLUSIONS

- A novel and versatile droplet experimental setup, developed at the Clark Atlanta University, was used to study the combustion and gasification behavior of hexane droplet at ambient pressure (~1 atmosphere). Because of its small size and compact design, the system can be used for both normal and microgravity tests in a ground station or in a microgravity environment.
- Free-falling non-combusting hexane and heptane droplets exhibited a  $d^2$ -law gasification behavior at sub- and supercritical experimental conditions. The gasification processes of the suspended droplets of different hydrocarbons were studied. The system pressure, temperature, and the molecular weight of the

hydrocarbon that formed the droplet had large effects on the gasification. At subcritical temperature, the maximum gasification rate occurred when the system pressure was close to the critical pressure. As the molecular weight of the hydrocarbon droplet increased, the gasification rate of the droplet decreased.

- The oscillatory deformation of a non-combusting free droplet of hexane at ambient pressure was enhanced as the system temperature increased. The acceleration of a free-drop droplet proportionally increased as the system temperature increased while the resistance force decreased.
- Buoyancy effect on the combusting droplet was more pronounced near and above the critical pressure. Under some conditions the falling combusting droplet reversed direction and floated upwards due to buoyancy effect.
- The video images of the hexane droplet behavior at temperatures close to the critical temperature showed that the droplet becomes dynamically unstable at temperatures close to 500 °C and disintegrates as a result of the vanishing surface tension as the critical temperature is reached.

### **PERSONNEL SUPPORTED**

The project supported Dr. Yaw D. Yeboah (PI), and Drs. Emmanuel Karikari and Zhicheng Wang (Research Scientists). In addition, seven undergraduate students listed below worked on the project as research assistants.

- Mr. Otis Threatt, Jr. graduated in May 1999 in Electrical Engineering and is now working with AT&T in Atlanta. Otis plans to go to graduate school after a few years of working.



- Ms. Tamara Gray graduated in Chemical Engineering in May 1999. Tamara worked at the US Patent Office after graduation. She is now employed by Milleken Corporation.
- Mr. Antron Palmer graduated in Chemical Engineering in May 1999 and is now employed with Milleken Corporation.
- Ms. Pamela Reid graduated in May 2000 and is now at Georgia Institute of Technology in Atlanta as a graduate student in Chemical Engineering.
- Ms. Adetola Abatan graduated in May 2001 and will be heading to the University of Pittsburg as a graduate student in Chemical Engineering in Fall 2001.
- Courtney Malbrue is a senior studying Chemical Engineering and Chemistry.
- Mr. Selasi Blavo is a Sophomore studying Chemical Engineering and Computer Science.

### **PUBLICATIONS**

Papers on the project are being prepared for publication in a reputable journal and the 29<sup>th</sup> International Symposium on Combustion.

### **PRESENTATIONS**

A presentation entitled "Study of the Sub- and Supercritical Behavior of Fuel Droplets" was presented at the AFOSR/ARO Contractors meeting in Santa Fe, New Mexico on June 12-14, 2000. A paper was accepted for presentation at the 1999 Technical Meeting of the Eastern States Section of the Combustion Institute, Rayleigh, NC, on October 12, 1999. The paper was entitled "Effects of Pressure, Molecular Weight

and Oxygen Potential on Hydrocarbon Droplet Gasification and Combustion Characteristics under Supercritical Pressure Environments.” Another presentation was given at the 1998 Technical Meeting of Central State Section of the Combustion Institute on June 1 at Lexington, Kentucky. The paper was entitled “Preliminary Study of the Droplet Gasification and Combustion Characteristics under Sub and Supercritical Pressure Environments.”

### **INTERACTIONS RELATED TO THE RESEARCH**

Joint experiments were carried out during this study with researchers at the Air Force Research Laboratory (AFRL) (Drs. Doug Talley and Bruce Chehroudi). The experiments were conducted by CAU and AFRL scientists in California using AFRL spray and jet research facilities and the CAU high-speed video imaging system. The results of the study were inconclusive.

### **INVENTIONS**

There was no invention from this research in the reporting period.

### **ACKNOWLEDGMENTS**

This work was supported by the Air Force Office of Scientific Research under a Grant No. F49620-98-1-0373). Especial thanks go to the Program Manager, Dr. Julian Tishkoff for his input, support and guidance and the AFRL Researchers, Drs. Doug Talley and Bruce Chehroudi, for their input and collaboration.

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